Coherent control of acoustic phonons in bulk semiconductors probed by time-resolved x-ray diffraction

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INTRODUCTION

The method of coherent control allows one to exploit classical and quantum-mechanical interferences to control the dynamics of atomic, molecular, and solid-state systems, and to drive them into novel, non-equilibrium states. This technique has been widely applied to control both electronic and vibrational degrees of freedom in a variety of systems[1,2]. Typically, a sequence of femtosecond pulses excites some arbitrary system into a state dependent on the temporal delay between pulses, and the resulting dynamics are probed optically. Here we use time-resolved x-ray diffraction with atomic-scale spatial resolution and picosecond temporal resolution to directly observe the coherently-controlled atomic motion as the solid moves along a particular vibrational pathway in which selected acoustic modes are either driven to large amplitude or cancelled out.

There are two complimentary ways of probing vibrational excitations in condensed matter through x-ray scattering. In the frequency domain, inelastic x-ray scattering with high energy resolution enables one to map out dispersion relations by resolving the change in energy of a photon scattered off a phonon at a given momentum transfer[3]. Alternatively, a phonon of wave-vector q modulates the diffraction efficiency of a crystal at a corresponding frequency ω. By resolving this effect on time-scales comparable to vibrational periods, the phonon frequency corresponding to a given q is directly measured, thus allowing the phonon dispersion relation to be recorded with picosecond temporal resolution[4].

There are two ways of viewing the control process used here. In the time-domain, a particular acoustic mode is excited twice, the second time with some chosen phase relative to the first. The first pulse starts up a coherent motion with all atoms moving in synchrony. The second pulse excites a phase-delayed replica of the first, and the two interfere constructively or destructively. Alternatively, one may view two-pulse excitation as pulse shaping of the impulsively-generated acoustic pulse, analogous to the shaping of femtosecond optical pulses in order to control, for example, quantum-mechanical transition probabilities[5].

EXPERIMENT

The setup is similar to previously reported experiments[4]. White light from a bending magnet beamline at the Advanced Light Source synchrotron is monochromatized in a two-crystal Germanium (111) monochromator to an energy of 5 keV with a spectral bandwidth of ~3 eV. A Ti:Al_2O_3-based 150 fs, 1 kHz, 800 nm laser, is synchronized to individual electron bunches within the storage ring with 5 ps jitter, much less than the ~60 ps electron bunch length. A Michelson interferometer with adjustable time delay splits the laser into two pulses which

impulsively excite acoustic phonons within the sample. The incident fluence for each pulse is 2 mJ/cm^2. The time-resolved diffracted intensity is then measured using a streak camera with 2 ps resolution in averaging mode, and triggered by a GaAs photo-conductive switch. The InSb sample is cut asymmetrically so that the [111] planes are oriented at an angle 16 degrees from the surface, and used in grazing incidence geometry so that the rocking curve of the (111) reflection is ~100 arcsec wide. This minimizes the finite range of phonon modes probed at each crystal angle and therefore increases the observed dephasing time of the acoustic oscillations. This dephasing time is then set by the time for the generated acoustic pulse to leave the x-ray probe region, rather than by the bandwidth from the monochromator or the intrinsic divergence of the synchrotron source. The acoustic pulses generated thus exhibit exhibit coherences which last on order 100 ps.

Fig. 1A shows the diffracted intensity (normalized to one for t<0) for single pulse excitation at an angle 300 arcsec from the (111) Bragg reflection, corresponding to the $q=4x10^5$ cm^{-1} mode. Following an initial drop in the diffracted intensity, large amplitude oscillations with a period of ~40 ps are observed, corresponding to the selected mode according to the linear dispersion relation $\omega=qv_{sound}$. Two pulse excitation creates, for small enough amplitudes to obey linear superposition, acoustic pulses of the form shown in the insets to Fig 1. A delay of 35 ps between the two optical pulses, equal to one vibrational period, results in constructive interference of the $q=4x10^5$ cm^{-1} mode and suppression of all nearby modes as shown in Fig 1B. In contrast, for a relative delay of 18 ps, or 1/2 of a vibrational period, the probed mode is completely silenced (Fig 1C).

CONCLUSION

Coherent control of acoustic phonons in a bulk semiconductor has been demonstrated and probed by time-resolved x-ray diffraction techniques. Two-pulse excitation enables one to drive one particular acoustic mode to large amplitude or to destructively cancel it out, thus accessing thermally inaccessible states of matter. The use of time-resolved x-ray diffraction to investigate structural phase transitions coupled with coherent control techniques offers the possibility of driving complex materials along a particular pathway into different structural phases of matter. Finally, the use of time-resolved x-ray scattering techniques to probe the coherently-controlled dynamics of matter on an atomic length-scale is the first step in the optimization of more complicated processes, in which the ideal control sequence is not yet known, but can be extracted through the direct observation, in real time, of the resulting atomic motion[6].

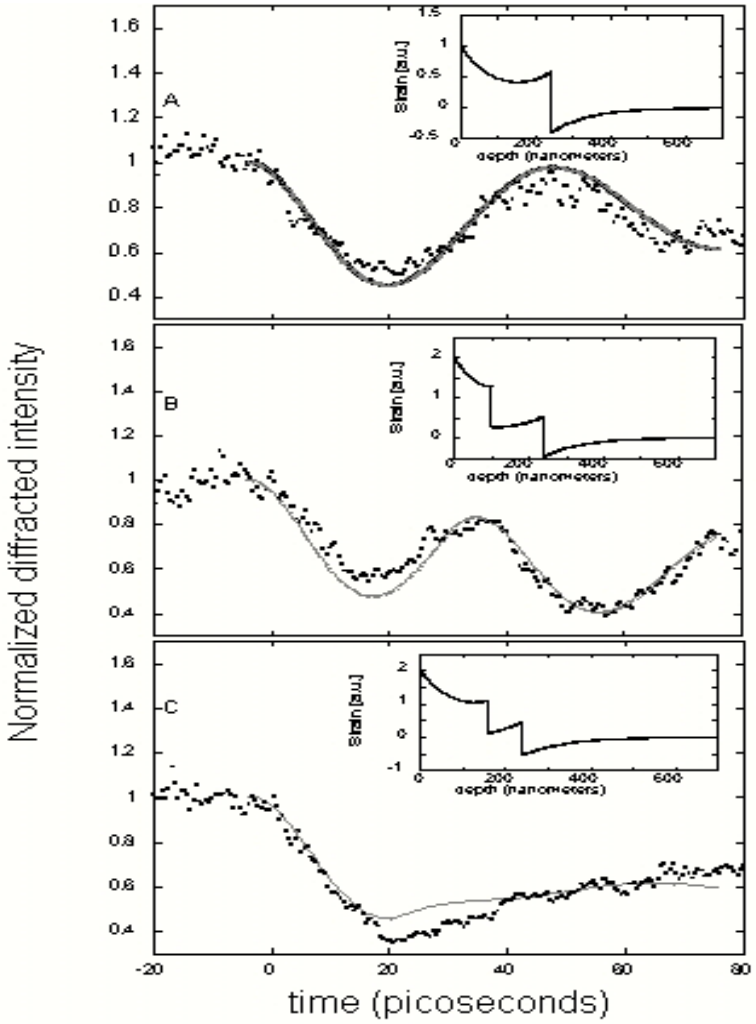

Figure 1. (**A**) Single pulse excitation, probing the $q=4x10^5$ cm^{-1} acoustic phonon mode. (**B**) Enhancement of the $q=4x10^5$ cm^{-1} mode using 2-pulse excitation with a relative delay of 35 ps, equal to one vibrational period. (**C**) Cancellation of the $q=4x10^5$ cm^{-1} acoustic using 2-pulse excitation with a relative delay of 18 ps, or 1/2 a vibrational period. Time-resolved diffracted intensity is normalized to one for $t<0$. Solid line corresponds to simulations. Insets: Generated acoustic pulse profile for each case, respectively, at time t=60 ps

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